

Entangling one polariton with a photon: effect of interactions on a single-polariton quantum state

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Polaritons are quasi-particles originating from the coupling of light with matter that demonstrated quantum phenomena at the many-particle mesoscopic level, such as BEC and superfluidity. A highly sought and long-time missing feature of polaritons is a genuine quantum manifestation of their dynamics at the single-particle level. Although they are conceptually perceived as entangled states and theoretical proposals abound for an explicit manifestation of their single-particle properties, so far their behaviour has remained fully accountable for by classical and mean-field theories. In this Article, we report the first experimental demonstration of a genuinely-quantum manifestation of microcavity polaritons, by swapping, in a two-photon entangled state generated by parametric down-conversion, a photon for a polariton. Furthermore, we show how single polaritons are affected by polariton-polariton interactions in a propaedeutic demonstration of their qualities for quantum information applications.

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Light would be the perfect platform for future quantum information processing devices, would it not be for its too feeble interaction. [1] A remedy is to rely on hybrid systems that involve a matter component, bringing in strong interactions. [2, 3] In the regime of strong coupling that binds together light and matter, the resulting polaritons appear as candidates of choices to deliver the strongly interacting photons required in tomorrow's quantum technology [4] and indeed, a photon-photon gate relying on these ideas has been recently demonstrated with a single atom in a cavity. [5] At a theoretical level, the polariton is itself an entangled superposition of light with a dipole-carrying medium, of the form $|U/L\rangle = \alpha |0_a 1_b\rangle \pm \beta |1_a 0_b\rangle$, with $|1_a\rangle$ a photon and, depending on the system, $|1_b\rangle$ a phonon, a plasmon, an exciton or even a full atom. The exciton-polariton, that lives in semiconductors, [6] has already demonstrated to be on par with cold-atoms physics, with reports that include Bose-Einstein condensation [7] (up to room temperature [8]), superfluidity, [9, 10] topological physics [11] and an ever growing list of other exotic quantum macroscopic phases. [12] This 2D-particle, that combines antagonist properties of light (lightweight and highly coherent) and matter (heavy and strongly interacting) as it propagates, also holds great promises for the future of classical technology by allowing the shift from electronic to optical devices [13] and emerging as the future generation of lasers. [14, 15] Thus, one of the major prospects of polaritons is their quest for quantum-based technologies, as quantum particles akin to photons but with much stronger nonlinearities.

Yet, even in configurations that are expected to provide strong quantum correlations, [16] exciton-polaritons have so far remained obstinately classically correlated. However useful is their description as a quantum superposition of excitons and photons, with α, β complex probability amplitudes, the actual state typically realized in the laboratory is instead a thermal or coherent distributions of such particles that yield, e.g., for the case of coherent excitation, product states $|\alpha\rangle_a |\beta\rangle_b$. [17] The variables α and β obey the same equations of motion and evolve identically to quantum probability amplitudes, but they now describe classical amplitudes of coherent fields for the photon (a) and exciton (b), with no trace of purely quantum effect such as entanglement and nonlocality.

This does not invalidate the interest and importance of reaching the genuinely quantum-correlated regime of polaritons [18–20] (from now on, we write polariton for exciton-polariton). Following decades of sustained efforts in this direction, the state-of-the-art is the recently reported unambiguous squeezing of the polariton field. [21, 22] This remains however a weak demonstration of non-classicality (a classical field can be squeezed in all its directions). What is instead required for a full exploitation of quantum properties is a stronger class of quantum states that are non

convex mixtures of Gaussian states, i.e., akin to Fock states and suitable to perform full quantum information processing. [23] Polariton blockade, [24] that relies on nonlinearities to produce antibunching, remained so far unsuccessful. Much hope was rekindled by the unconventional polariton blockade [25, 26] based on destructive interferences of the paths leading to two-excitation states, allowing much stronger antibunching, although not in the sought class beyond convex mixtures of Gaussian states. [27] The race for quantum polaritonics is thus still in its starting blocks and several questions remain pending to justify polaritons as strong contenders for quantum information processing. Indeed, one of the main uncertainties in the community is whether polaritons are robust against pure dephasing. Being composed of interacting particles—excitons—they could be strongly affected by the environment in a harmful way for their quantum coherence. Furthermore, even assuming that one could generate a polariton qubit, another wishful quality of polaritons remains to be established: can a single polariton be substantially affected by interactions with other polaritons? This is of paramount importance to use polaritons as building blocks for nonlinear quantum gates.

In this Article, we answer positively to these two outstanding questions, by turning to a new paradigm for creating quantum polaritons. Instead of realizing a genuine quantum state from within, e.g., based on strong nonlinearities, we imprint it from outside. [28] Since photonics is so-far leading in the generation, transfer and manipulation of quantum states, and since photons couple well to polaritons, this opens an opportunity to realize and explore quantum effects with polaritons, leaving to technology to provide self-serviced samples in the future. To ensure the quantum nature of the polariton field, we use entangled photon pairs: one photon is sent to the microcavity and the other is used to later check that quantum correlations are still present and, therefore, have been transferred to the polaritons. The source of entangled photon-pairs is a continuous wave (cw) laser at $\lambda = 405$ nm, down-converted from a Periodically Poled KTP crystal (PPKTP) [29] in order to generate pairs of photons with a bandwidth narrow enough to couple to our microcavity. The latter is described in the Methods. The PPKTP is introduced inside a Sagnac interferometer [29] (see sketch in Fig. 1) which allows to create polarization entangled photons in a state of the form $|\Psi\rangle = (1/\sqrt{2})(|HV\rangle + e^{i\phi}|VH\rangle)$, where $|H\rangle$ stands for a horizontally polarized photon and $|V\rangle$ for a vertically polarized one. The phase can be controlled to create a Bell state $|\Psi^\pm\rangle = 1/\sqrt{2}(|HV\rangle \pm |VH\rangle)$ by placing a liquid crystal in one arm of the interferometer, at the end of which a pair of non-degenerate entangled photons is emitted in different directions: the idler at the wavelength of the polariton resonances ($\lambda \sim 830$ nm), and the signal at higher energies ($\lambda \sim 790$ nm). The high-energy photon is then directed to a standard polarization to-

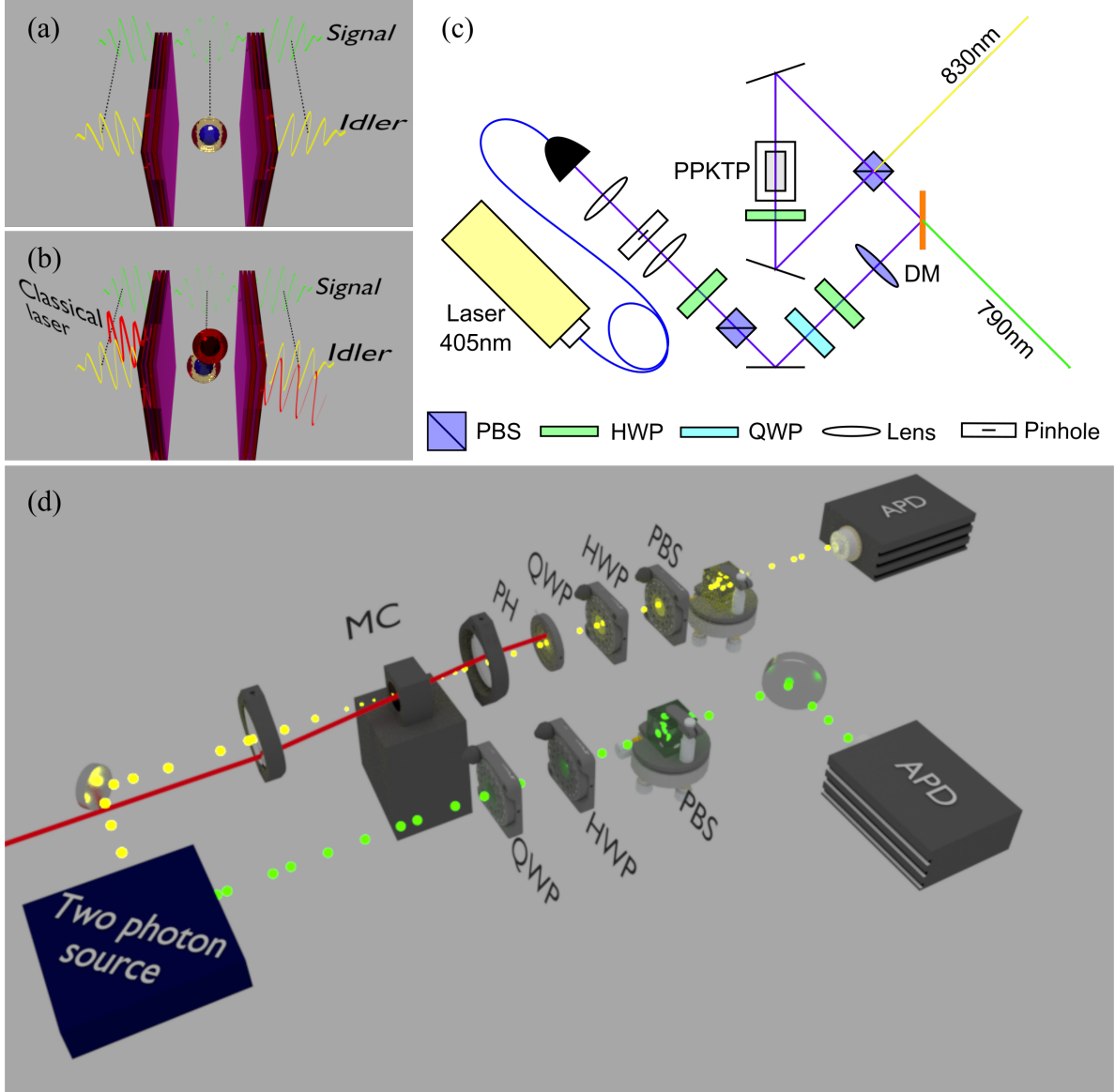


FIG. 1. Sketch of the behaviour inside the microcavity: (a) Linear regime: a single photon (yellow, idler) gets into the microcavity where it becomes a polariton, that is later reconverted into an external photon. When this photon is entangled with another one (green, signal), we show that the entanglement is preserved throughout. (b) Interacting regime: a single photon (yellow, idler) enters the microcavity along with photons from a classical laser (red). The polaritons in the microcavity interact. This affects the entanglement of the single-polariton in a way that allows to measure the polariton-polariton interaction. (c) Sketch of the Sagnac interferometric source. (d) Sketch of the setup implementing these configurations.

mography stage, consisting of a quarter waveplate (QWP) followed by a half waveplate (HWP), a polarizing beamsplitter (PBS) and a single-photon detector (APD). The low-energy photon, instead, is directed towards the microcavity where it excites a single polariton, is stored there until its eventual re-emission, is retrieved and finally directed to a second tomography stage, equal to

the one for the first photon. The coincidence counts from the two single-photon detectors are measured in a 4 ns window with a home-made coincidence unit. By making such measurements for all the combinations of polarizations on each detector (that makes 6×6 possibilities for the three polarization bases: circular, vertical and diagonal), [30] we are able to determine whether the photon that transformed into a polariton and back retained the nonlocal quantum correlations of a Bell state. If this is the case, this proves that the polariton state that inherited and later passed this information was itself in a genuine one-particle quantum state, and was furthermore entangled with the external photon propagating on the other side of the setup. Namely, we have created the state $|\Psi^\pm\rangle = (1/\sqrt{2})(a_H^\dagger p_V^\dagger \pm a_V^\dagger p_H^\dagger)|0\rangle$ where p_H and p_V are the boson annihilation operators for the horizontally and vertically polarized polaritons and a_H and a_V for the signal photon. While this could be well expected from a standard linear theory of light propagating through resonant dielectrics, the failures of polaritons to demonstrate other expected quantum correlations, such as in OPO scattering, show that it is not at all obvious that polaritons are proper carriers of quantum features. In particular, their composite nature and large exciton component could lead to fast decoherence (e.g., via phonon scattering).

The spectral shape of the idler state is shown in Fig. 2(a). Using single mode laser excitation of the PPKTP crystal, the bandwidth of the entangled photon pairs is reduced to 0.46 nm, which is only 35% wider than the polariton state. Using temperature tuning on the nonlinear crystal we could move the idler resonance from 825 nm to 831 nm. In order to check that the entangled idler is transferred into the polariton state and not passing through the cavity mode, we performed transmission measurements while scanning the energy of the idler from below the lower polariton branch (LPB) to above the upper polariton branch (UPB). In Fig. 2(b), the effect of the microcavity on the idler state shows that no light is transmitted when the idler is out-of-resonance with the polaritons. This means that every photon that passes through the sample has been converted into a polariton. If this would not be the case, we would have observed at the cavity mode (between the LPB and UPB) a finite signal, which is completely absent even in logarithmic scale (not shown).

For a bipartite system such as our photon pair, entanglement can be quantified through the concurrence, defined in the Methods, which is zero for classically correlated or uncorrelated states and gets closer to one the higher the quantum entanglement. To study the transfer of entanglement into the polariton field, we compare the concurrence of the two photons without the microcavity (corresponding to the maximally entangled state) to that in which the photon is emitted by the microcavity after the idler photon has been converted into a polariton of the LPB with zero momentum ($k_{\parallel} = 0$). As can be seen in Fig. 3, the concurrence diminishes from 0.826 in the case

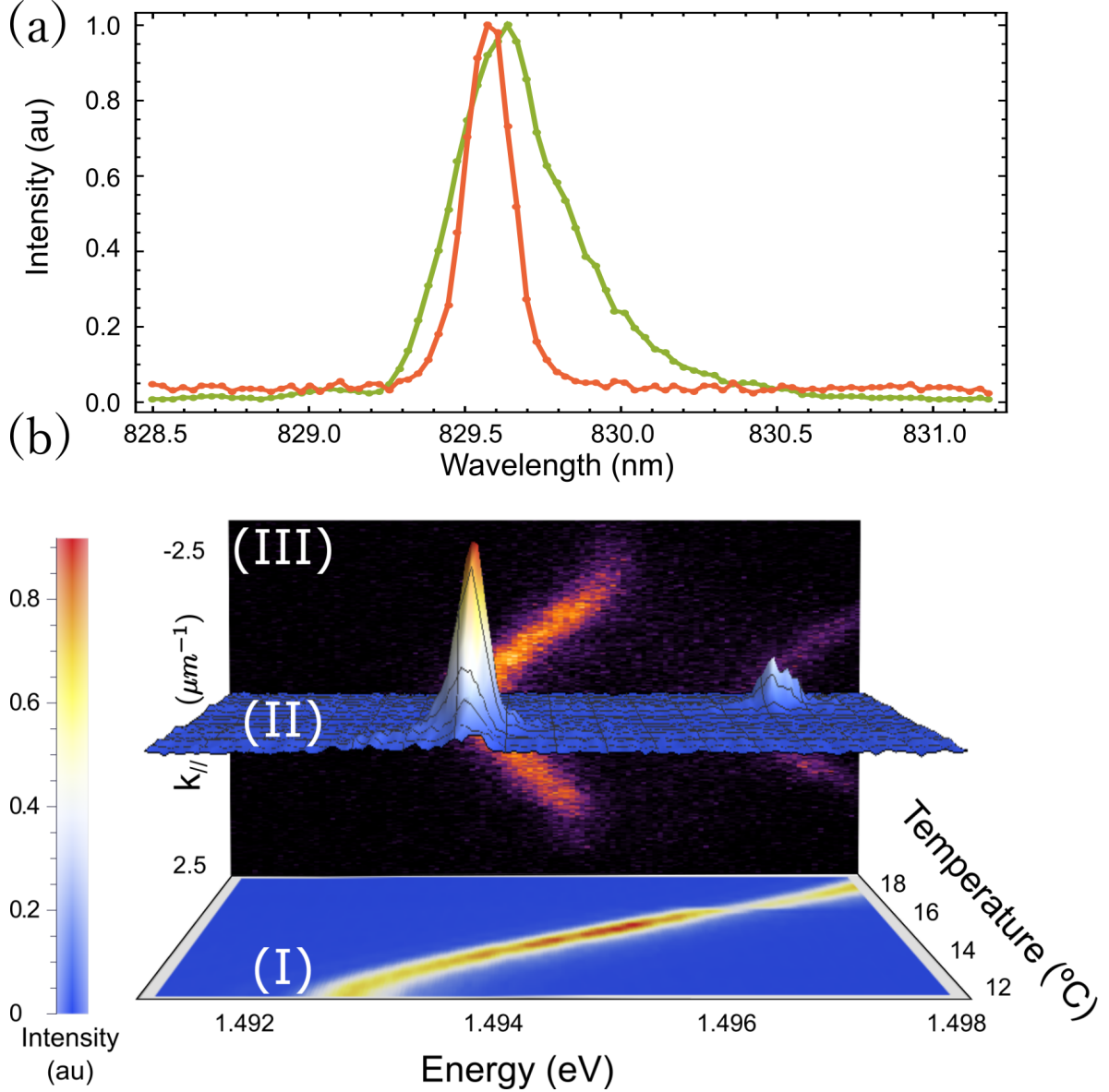


FIG. 2. (a) Green: normalized emission of the PPKTP crystal. Orange: normalized transmission of the emission of the PPKTP crystal through the LPB. (b) Changes on the resonance as a function of the temperature of the two-photon source. I: 2D map of the emission of the crystal outside of the microcavity. II: transmitted intensity as a function of the energy as the temperature varies. Two peaks can be identified that correspond to the resonances with one of the two polariton branches. The color-bar corresponds to both I & II. III: far field of the emission under non-coherent pumping.

of two freely-propagating photons, to 0.806 when one of the photons is converted into a polariton. The maximum concurrence of the PPKTP source is limited to 0.826 because the entangled state could not be optimized at the operation wavelengths needed to interface photons with polaritons. Nevertheless, the concurrence is large and remains so in presence of a microcavity in the way. This

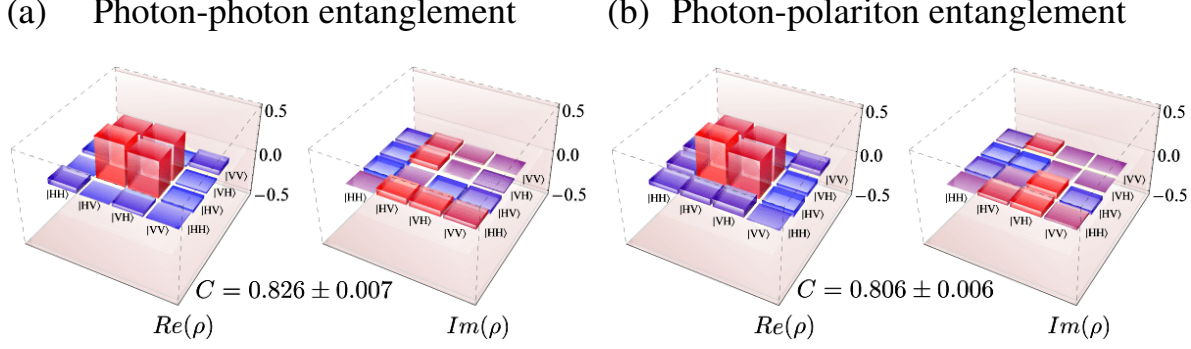


FIG. 3. Tomography measured between the signal and idler photons. The signal is sent directly towards the detector while the idler photon becomes a polariton when entering the sample. (a) Real (left) and imaginary (right) components of the density matrix for the source of photon-pairs without the sample. The concurrence is not unity because the operation wavelength is not optimal for the source. (b) Real (left) and imaginary (right) components of the density matrix when passing through the microcavity. The concurrence of 0.806 shows that polaritons retain the entanglement.

result is a direct proof that we have generated a single polariton within the microcavity and that it existed there as a quantum state with no classical counterpart. If the single photon was lost in the polariton thermal noise or coupled to a collection of other polariton states, no entanglement would be eventually observed. This transfer of the photon to and from a polariton state has shown to conserve almost entirely the original degree of entanglement. Such a result is encouraging for a future exploitation of quantum polaritonics.

Beyond the preservation of concurrence, we also demonstrated the nonlocality between the signal photon and the polariton created by the idler photon. We did so by probing the classical Clauser–Horne–Shimony–Holt (CHSH) inequality [31] $S \leq 2$ where:

$$S = |E(a, b) - E(a, b') + E(a', b) + E(a', b')|, \quad (1)$$

with

$$E(x, y) = \frac{C_{++}(x, y) + C_{--}(x, y) - C_{+-}(x, y) - C_{-+}(x, y)}{C_{++}(x, y) + C_{--}(x, y) + C_{+-}(x, y) + C_{-+}(x, y)}, \quad (2)$$

a function of the coincident counts $C_{pq}(x, y)$ between the two measurement ports of our tomography setup, for the photons with polarization $p, q \in \{-, +\}$ in the bases $x = a, a'$ and $y = b, b'$ where $a = -\frac{\pi}{8}$, $a' = \frac{\pi}{8}$, $b = 0$ and $b' = \frac{\pi}{4}$ are the combinations of polarization angles that maximize the Bell's inequality violation. This can be obtained by rotating the tomography HWPs in $a/2$, $a'/2$, $b/2$ and $b'/2$. Figure 4 displays the measured Bell curves, which show the photon coincidences associated to these bipartite polarization measurements, along with the continuous correlated oscillations

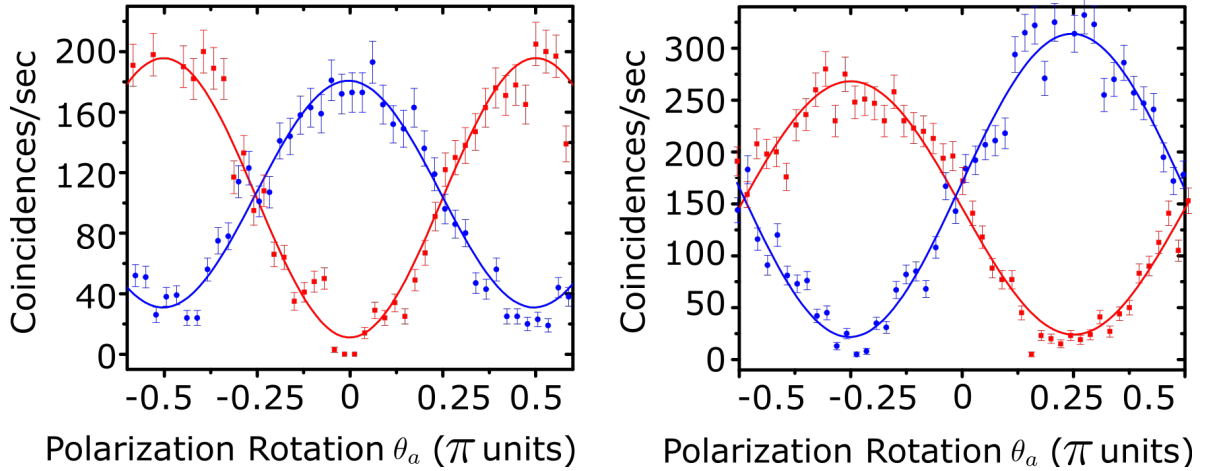


FIG. 4. Coincidences as a function of the polarization between the external photons and the polaritons. Left: Bell curves for a polarization angle $b = 0$. Right: Bell curves for $b = -\pi/4$. Red squares and blue circles denote the $++$ and $+ -$ coincidences, respectively.

predicted by the theory. In the optimum configuration, we obtain a value of $S = 2.463 \pm 0.007$ which unambiguously violates the CHSH inequality and proves the nonlocal character of the photon-polariton system.

The above experimental results clearly demonstrate that, although polaritons are quasi-particles in a solid-state system with complex and yet-to-be-fully-characterized interactions with their matrix and other polaritons, [32] they can be used as quantum bits maintaining almost unvaried their quantum state and can transfer it back and forth to an external photon. In particular, this shows that several effects, such as pure dephasing, coupling to phonons, radiative lifetime, etc., are not detrimental to quantum coherence.

To answer the second important question on the possibility to affect the quantum state that we have created, we need to go into the nonlinear regime and study the effect of polariton interactions. To observe such nonlinearities within our available equipment, we repeat the experiment in a non polariton-vacuum configuration. Namely, instead of exciting the sample with one photon of the PPKTP source only, we add the excitation of a classical laser, which is the most common way to excite a microcavity (see sketch in Fig. 1). In this case, the entangled photon is sent on resonance now with the UPB, and the classical laser with the LPB. We have chosen this configuration to avoid any effect of relaxation of the classical source into lower energy states while at the same time keeping the quantum state well distinguishable from the other polaritons.

The classical pumping power is changed from having only the vacuum state up to an average of 230 polaritons at any given time, with a density still below $1 \text{ polariton}/\mu\text{m}^2$. The calibration of

the population is explained in the Methods.

In the conditions of our experiment, the full interacting-polariton Hamiltonian can be reduced to the simple form (See Supplementary Material):

$$H = \omega_{\uparrow} q_{\uparrow}^{\dagger} q_{\uparrow} + \omega_{\downarrow} q_{\downarrow}^{\dagger} q_{\downarrow} + g_{\uparrow\downarrow} (q_{\uparrow}^{\dagger} q_{\downarrow} + q_{\downarrow}^{\dagger} q_{\uparrow}), \quad (3)$$

where q_p are quantized operators for the upper polariton at $k = 0$ with polarization $p = \uparrow, \downarrow$ and the lower polariton condensate has been absorbed in the coefficients through a mean-field approximation for the coherent state $|\alpha_{\uparrow/\downarrow}\rangle$ with polarization \uparrow / \downarrow :

$$\omega_{\uparrow/\downarrow} = \tilde{\chi} (3|\alpha_{\uparrow/\downarrow}|^2 V^{(1)} + 2|\alpha_{\downarrow/\uparrow}|^2 V^{(2)}) + \omega_0, \quad (4a)$$

$$g_{\uparrow\downarrow} = 2\tilde{\chi} |\alpha_{\uparrow}\alpha_{\downarrow}| V^{(2)}, \quad (4b)$$

where $\tilde{\chi} \approx 0.2$ and ω_0 are some constants linked to the Hopfield coefficients that arise due to the geometry of the experiment (see Supplementary) and, more importantly, $V^{(1,2)}$ correspond to same (1) or opposite (2) spin polariton-polariton interactions. The nonlinear crystal emits pair of polarization-entangled photons of the form:

$$|\psi_0\rangle = \frac{1}{\sqrt{2}} (|H, V\rangle + |V, H\rangle) \equiv \frac{1}{\sqrt{2}} (c_H^{\dagger} q_V^{\dagger} + c_V^{\dagger} q_H^{\dagger}) |0\rangle, \quad (5)$$

where c_p is the quantum operator for the signal photon that goes straight to the detector. The q_p photon, on the other hand, evolves according to the Hamiltonian (3). Even for the free propagation $q_p^{\dagger} q_p$, the possible asymmetry for the $p = H$ and V polarizations results in different phase-shifts, which alter the wavefunction as a whole. The rightmost term in Eq. (3), on the other hand, results in a change of the state of polarization. By analyzing the quantum correlations between the signal and idler after passing through the cavity, one can thus gain information on the microscopic parameters $\omega_{\uparrow/\downarrow}$ and $g_{\uparrow\downarrow}$.

At the level of the concurrence, our observation is unambiguous. As shown in Fig. 5 (data points), the concurrence decays with increasing polariton density (following increasing laser pumping). While the evolution of the wavefunction is expected, the loss of concurrence seems to suggest a decoherence of the single polariton when affected by interactions with the condensate. However, this is due to a different scenario, beyond mean-field effects of the lower polariton condensate, which, not being a Fock state, has fluctuations (Poissonian ones) [33]. While in absence of fluctuations, the model predicts that the state of the entangled polariton acquires a relative phase-shift between the spin states of the superposition (See Supplementary Material), which causes no loss of concurrence (dashed green line), the averaging over several pairs with a fluctuating phase-shift,

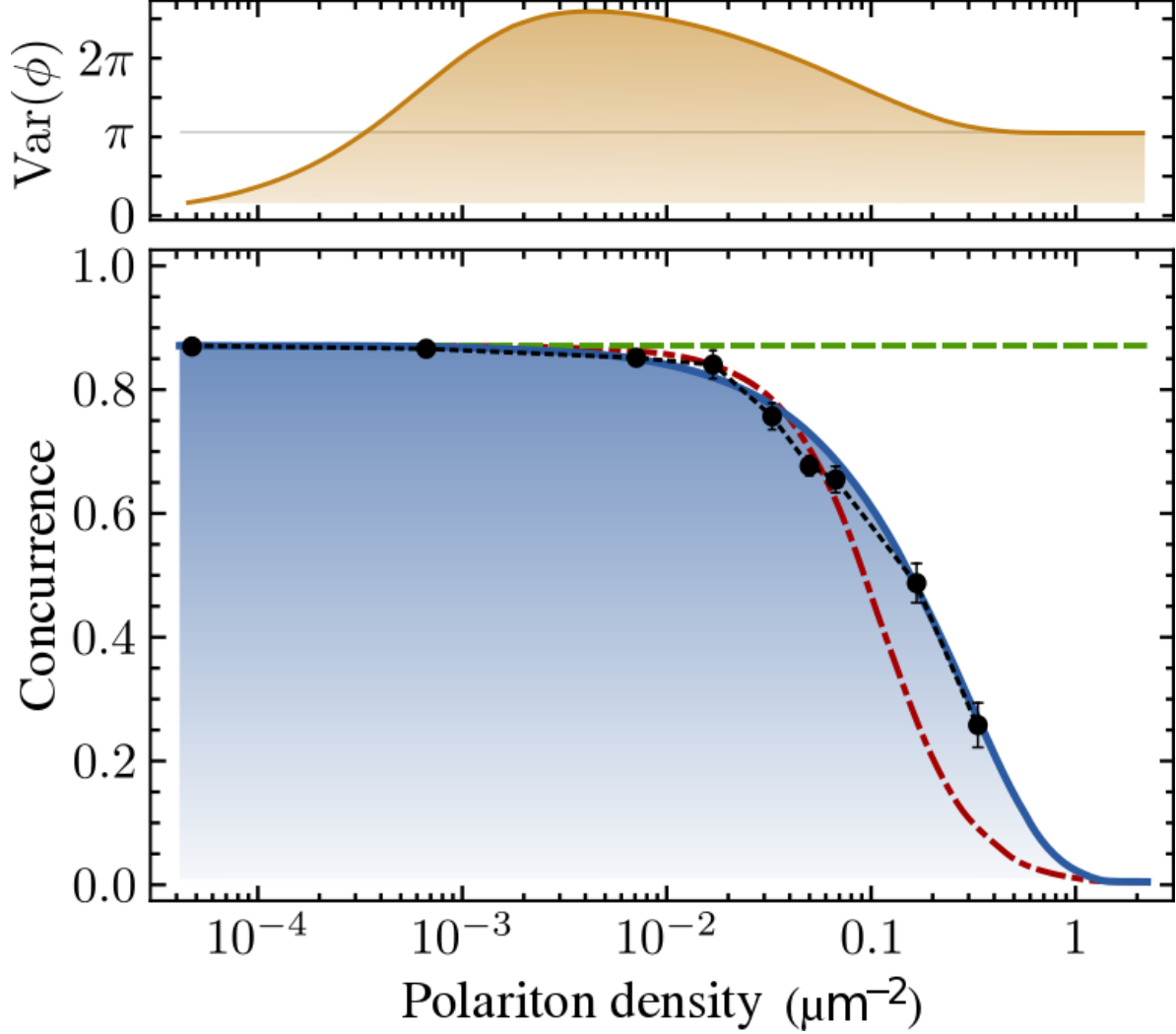


FIG. 5. Concurrence \mathcal{C} between the external photons and the polaritons as a function of the mean number of polaritons present in the sample (horizontal axis) from a classical laser. Each data point is obtained from the 36 measurements of coincidences in all the combinations of polarization (dotted line serves as a guide). The solid line is a theoretical simulation for a model of fluctuating polarized lower polaritons that interact with the single upper polariton injected by the quantum source with an interaction strength of 16% of the radiative broadening, and which is in excellent quantitative agreement with the observation. The orange line shows the variance of the phase-shift due to the fluctuations of the lower polaritons. The variance reaches its maximum when phase-shifts start to be stronger than 2π , thus causing a small dephasing. In the limit of very large fluctuations, the variance of the phase is that of a uniform distribution between 0 and 2π , namely $\sigma^2 = \pi^2/3$ (thin horizontal gray line). The green, dashed line is the theoretical model without the fluctuations, whereby the concurrence remains unaffected by the polariton condensate. The red, dashed-dotted line shows the theoretical simulation for a model of a condensate assuming thermal fluctuations, which fails to fit quantitatively the experimental observation and confirm that the condensate has Poissonian fluctuations, as expected.

concomitant with the fluctuating condensate population, has the effect of an effective decoherence. In this case, each entangled pair sees the microcavity in an instantaneous different state, evolving the wavefunction differently and acquiring a different relative phase-shift, which results in spoiling the entanglement *on average* as measured through the signal/idler correlations in polarizations. Implementing this effect in the model allows us to reproduce theoretically the experimental findings with an excellent agreement (solid blue line). We also show the spread of the relative phase (orange dashed line) as measured by its variance, that confirms that concurrence is lost due to a fluctuating phase-shift, since the concurrence starts to decay when the variance has reached its maximum, indicating that the phase is randomly distributed (the variance decays because the phase-shift is bounded modulo 2π ; when fluctuations are so strong as to make the phase-shift uniformly distributed, the variance becomes $\pi^2/3$ and the concurrence vanishes). This shows that the wavefunction of a single-polariton Fock state is strongly affected indeed by its interactions with the condensate in the LPB, albeit in a random way as ruled by fluctuations currently beyond our control. However, in future experiments, if the condensate would be replaced by another non-fluctuating quantum state, such as another Fock state, this would lead, instead of an apparent decoherence, to a well-controlled spin-dependent phase-shift that would be able to power quantum circuits. In fact, if the lower polariton branch would be in a Fock state $|n_\uparrow, n_\downarrow\rangle$ with $n_{\uparrow,\downarrow}$ polaritons with polarization \uparrow, \downarrow , respectively, the model shows, neglecting the smaller $V^{(2)}$ (see Supplementary Material) that a single polariton in the upper polariton branch would then acquire a relative phase-shift of:

$$\phi = 3V^{(1)}\tau\bar{\chi}(n_\downarrow - n_\uparrow), \quad (6)$$

which, thanks to the large exciton-exciton repulsion $V^{(1)}$, results in a sizable shift, namely, with circularly-polarized lower-polaritons only and for the parameters fitting the experiment, $\phi \approx \frac{\pi}{30}n_\downarrow$. With polaritons of 60 ps lifetime, which is well within the state-of-the art, one would thus get a deterministic π -shift for the single polariton in the upper branch induced by one polariton only in the lower branch, which would allow the realization of a polariton controlled-Z gate, the fundamental building block for a controlled-NOT (CNOT) gate. Our results are thus promising for on-chip implementation of nonlinear quantum gates.

One can go even further in characterizing the underlying Hamiltonian by mapping its effect on all the possible polarized qubit states, i.e., how it transforms the Poincaré sphere. When in possession of entangled states, as in our case, this can be achieved with a single input state only, thanks to a technique (further detailed in the Methods) known as ancilla assisted quantum process

tomography. [34] In essence, the sphere rotates under unitary transformation and shrinks under the effect of decoherence. Our joint experimental/theoretical analysis, presented in Fig. 6, shows that the sphere does not rotate at lower pumping, although it experiences some wobbling, until, at larger pumping, it shrinks into a spindle, whose main axis does rotate. The experimental tomography is a direct post-processing of the data. The theory applies the same procedure to input states (5) undergoing the evolution of Hamiltonian (3). We find that the shrinking into a spindle, also due to the fluctuating condensate that causes the loss of concurrence, can be obtained with no rotation of the sphere with increasing pumping when $\omega_{\uparrow} - \omega_{\downarrow}$ and $g_{\uparrow\downarrow}$ remain constant on average. In these conditions, both polarization states at the extremities of the spindle have a constant energy shift and there is no admixture of polarization. This results in decohering all polarization states except in the \uparrow, \downarrow basis, while not producing a unitary rotation. The fact that the spindle shrinks along an axis that varies with power suggests that at higher pumping, the polariton condensate renormalizes itself into a condensate of polarized quasi-particles in an arbitrary x, y polarization basis, so that the effective Hamiltonian for the upper polariton polariton becomes $H = \omega_x q_x^{\dagger} q_x + \omega_y q_y^{\dagger} q_y + g_{xy} q_x^{\dagger} q_y + g_{xy}^* q_y^{\dagger} q_x$. This introduces new microscopic parameters, $\omega_{x/y}$ and g_{xy} , which, when kept constant in average with increasing pumping but still fluctuating, reproduce the experimental finding (cf. Fig. 6). The details of the microscopic origin of these terms are given in the Supplementary material. They are a complex admixture of the interaction strengths and the state of the condensate which in principle, could also be studied for their own sake, although this is out of reach of the present work. This shows, however, that the technique also opens new perspectives for probing classical features of the polaritons, such as the nature of the polariton condensate and of its interactions, known to depart from the atomic paradigm. [35]

In conclusions, the observation of a genuinely quantum state of the polariton field has been demonstrated. Our experiments also bring to semiconductors an implementation of quantum spectroscopy [28, 36–38] by using quantum-correlated light to access phenomena and information out of reach of a classical laser. Our measurement in the presence of a polariton condensate is the first of its kind and already confirms that polaritons are serious players on the quantum scene through their ability to exhibit strong nonlinear effects involving a single-polariton state. This has obvious implications for the design and implementation of a new generation of quantum gates, routing strongly-interacting polaritons in pre-determined landscapes so as to make them interfere [39]. Our experiment might well be the precursor for the long-sought strongly interacting photons needed for the realization of scalable and efficient quantum computers.

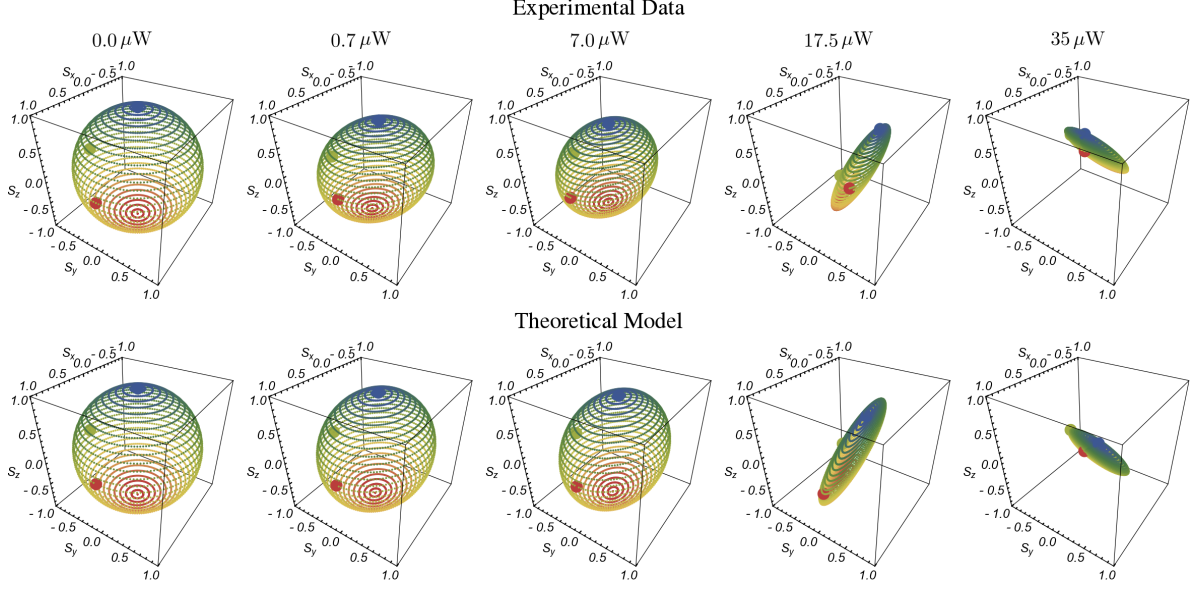


FIG. 6. Ancilla assisted quantum process tomography. Our technique allows us to observe the effect of the lower polariton condensate on the polarization of an upper polariton qubit, as a function of the increasing pumping (upper row). The shrinking of the Poincaré sphere into a spindle confirms that the qubit is affected by its interaction with the lower condensate, whose fluctuations lead to an effective decoherence when averaging. The fact that the decoherence occurs along a main axis x - y suggests that the condensate experiences a renormalization of its state in this particular basis. Making this assumption in the theoretical model leads to an excellent agreement of the observed phenomenology (lower row) and points at future methods for characterizing also the condensate itself. The figures for the theoretical model were obtained for 0, 0.018, 0.032, 0.226 and 0.272 polaritons per micrometer squared, respectively.

METHODS

A. Theoretical model

The theory models the experiment by feeding Hamiltonian (3) (see Supplementary for its derivation from the full polariton Hamiltonian) with the initial state (5) and acquiring statistics over repetitions of this scenario with coefficients (4) fluctuating with Poissonian distributions with a mean $|\alpha_{\uparrow\downarrow}|^2$, modeling a polariton condensate beyond mean-field.

The entanglement for bipartite systems can be quantified unambiguously with the “concur-

rence”, which extracts the amount of non-classical correlations from an averaged density matrix:

$$\bar{\rho} \equiv \frac{1}{\mathcal{N}} \begin{pmatrix} \text{HHHH} & \text{HHVH} & \text{HHHV} & \text{HHVV} \\ h.c. & \text{HV VH} & \text{HVHV} & \text{HV VV} \\ h.c. & h.c. & \text{VHHV} & \text{VHV V} \\ h.c. & h.c. & h.c. & \text{VV VV} \end{pmatrix}, \quad (7)$$

where \mathcal{N} is a constant put here so that $\text{Tr}(\bar{\rho}) = 1$ and, e.g., VHHV stands for:

$$\frac{1}{N} \sum_{j=1}^N \langle \psi_j | c_V^\dagger q_H^\dagger q_{HC}^\dagger | \psi_j \rangle, \quad (8)$$

where the states $|\psi_j\rangle$ are those computed from the fluctuating Hamiltonian as explained in the Supplementary Material. This corresponds to the experimental configuration where every element of the density matrix is reconstructed through a tomographic process that requires up to 36 measurements detecting every possible combination of polarization. From the density matrix in Eq. (7) we compute the concurrence as $\mathcal{C}[\bar{\rho}] \equiv \max(0, \lambda_1 - \lambda_2 - \lambda_3 - \lambda_4)$ where the λ_i are the eigenvalues in decreasing order of the matrix $\sqrt{\sqrt{\bar{\rho}} \tilde{\rho} \sqrt{\bar{\rho}}}$. Here $\tilde{\rho} \equiv (\sigma_y \otimes \sigma_y) \bar{\rho}^T (\sigma_y \otimes \sigma_y)$, and σ_y is a Pauli spin matrix.

The *Ancilla Assisted Quantum Process Tomography* (AAQPT) characterises the action of an unknown map (black box) acting on one of two entangled qubits, by using only one input and one output state to recreate the associated map. This is thanks to the intrinsic correlations of the bipartite state that dispense from considering the transformation of a large collection of input states.

Since any qubit $|\psi\rangle$ encoded in the polarization degree of freedom has a Stokes decomposition of its associated density matrix $\rho = \frac{1}{2} \vec{r} \cdot \vec{\sigma} = \frac{1}{2} (r_0 \mathbf{1} + r_1 \sigma_1 + r_2 \sigma_2 + r_3 \sigma_3)$ where \vec{r} is the Stokes vector and $\vec{\sigma}$ is the vector of Pauli matrices, [30] AAQPT allows to obtain the black box (the microcavity with a lower polariton condensate in our case) map χ from the expression $\vec{r}_{\text{out}} = \chi \vec{r}_{\text{in}}$ where \vec{r}_{in} and \vec{r}_{out} are the Stokes parameterization of the state of the single photon before and after it goes through the cavity, respectively. The numerical calculation of χ is obtained by the linear decomposition $\chi = (A^{-1}B)^T$ with A and B obtained from the state of the pair of entangled photons before and after one of the photons has gone through the cavity and are defined as $A_{i,j} = \text{Tr}[(\sigma_i \otimes \sigma_j) \rho_{\text{out}}]$ and $B_{i,j} = \text{Tr}[(\sigma_i \otimes \sigma_j) \rho_{\text{in}}]$. In the experiment, where errors and noise could lead to a non-physical result, the map is best estimated by a Maximum Likelihood process over χ . This technique minimizes the statistical dispersion of the expectation values between χ_{raw} and its trace preserving and completely positive version χ_{physical} . An intuitive representation of the map χ that

facilitate its interpretation is given through the Poincaré sphere, where any qubit is mapped as a point on the surface, using (r_1, r_2, r_3) as (x, y, z) coordinates (Fig. 6). There, r_0 represents the intensity of the field, r_1 the degree of linear polarization, r_2 that of diagonal polarization and r_3 that of circular polarization.

B. Sample & Setup

The microcavity sample is composed of front and back Distributed Bragg Reflectors (DBR) with 20 pairs each, confining light, and one $\text{In}_{0.05}\text{Ga}_{0.95}\text{As}$ quantum well (QW), confining excitons. The QW is placed at the antinode of the cavity to maximize their interaction and enter the strong-coupling regime. [40] The experiment consist in four different parts: photon generation, signal-photon tomography stage, polariton source and polariton tomography stage. The first part consists of a Sagnac interferometer excited with a single-mode diode laser at 405 nm and a pumping power of 6.5 mW and bandwidth FWHM < 5 pm. Its power selection is achieved by fixing a half-wave plate (HWP) before a polarizing beam splitter (PBS), and the horizontal output polarization is again rotated in a desired arbitrary polarization before a dichronic mirror (DM). In order to increase the efficiency of the PPKTP, a lens is introduced to focalize the diode right on the crystal. The second and the fourth sections of the setup are used for the tomography measurement. The idler photon goes through the polariton source, which consists on a cryostat at 20 K and a pressure of 100 mbar. The single-photon is focused on the surface of the microcavity and the emitted photons are recollected with a second lens sending them directly to the fourth stage (the idler's tomography stage). In the experiment measuring nonlinear effects, an additional laser is sent to the microcavity at a particular angle. The transmitted photons given by the laser are covered with a diaphragm (pin-hole, PH) at the Fourier plane of the recollection lens. The entanglement measurement takes place in the tomography analysis. [41] In our case, we apply a hypercomplete tomography by projecting the bipartite state onto a combination set of three bases: logical ($|\text{HVHV}\rangle$ & $|\text{V}\rangle$), diagonal ($|+\rangle = |\text{HVHV}\rangle + |\text{V}\rangle$ & $|-\rangle = |\text{HVHV}\rangle - |\text{V}\rangle$), and circular ($|R\rangle = |\text{HVHV}\rangle + i|\text{V}\rangle$ & $|L\rangle = |\text{HVHV}\rangle - i|\text{V}\rangle$). Each local projection is done by applying a rotation in a QWP and a HWP, followed by a PBS as polarization filter (see Fig 1). Finally, the remaining photons belonging to the qubit of both the signal and the idler are coupled to a single-mode optical fiber (SMF), connected to an avalanche photo-detector (APD). The reconstruction of the state sees the relative coincident count events (CC) among all the mentioned projections during the desired integration time. In that way, we measure the state by projecting many copies of the same. The measurements

corresponding to the results reported in Fig. 5 were unavoidably affected by the presence of the classical cw laser which was minimised by momentum selection of the polariton signal (as shown in Fig. 1d). The contribution of this noise, however, was subtracted from the raw data by performing desynchronised tomography for each power of the external laser. The calibration of the population and density in presence of the classical laser was obtained by means of four different parameters: the pumping power, the photon energy, the polariton lifetime and the laser spot size. The amount of photons/sec delivered by the laser to the microcavity were calculated as the ratio Power/Energy. Given the transmission of the microcavity and assuming the same amount of photons to be emitted on both sides of the sample, only 0.5% of the photons become polaritons. Multiplying this rate of polaritons/sec by the lifetime of a polariton (measured as 2 ps), gives a maximum of 230 polaritons for the highest power used. The polariton density polaritons was obtained by dividing by the spot area ($706 \mu\text{m}^2$).

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AUTHOR CONTRIBUTIONS

DS and FPL conceived the idea. AC, BSF, MD, FC and DGS prepared the set up and performed the experiments with help from DB and LD. VT, GB grew the sample and AF prepared the sample for transmission measurements. JCLC, CSM, EdV developed the theory under the supervision of FPL. LM, PM and FS supervised the experiments on photon-photon and photon-polariton concurrence. DS coordinated and supervised the project. All authors contributed to the analysis and interpretation of the data as well as the editing of the manuscript.

Authors declare no financial competing interest.

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